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Shapes of InAs quantum dots on InGaAs/InP

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InAs self-assembled quantum dots were grown on InGaAs lattice-matched on InP by metalorganic chemical vapor deposition. The facet formation on the dot was investigated by atomic force microscopy and transmission electron microscopy. The {136}-faceted InAs dots were elongated along either $[1\bar{3}0]$ or $[\bar{3}10]$ to form parallelogram-shaped islands analogous to hut cluster formation in SiGe/Si quantum dots. Some parallelogram dots also exhibited {110} faceting, presumably on undergoing a shape transition toward dots with facets of higher symmetry. © *2004 American Institute of Physics*. [DOI: 10.1063/1.1840123]

Self-assembled quantum dots (SAQDs) in heteroepitaxial systems have recently been the subject of intense research, because they can be applicable for zero-dimensional structures that have potential applications in future high-performance electronic and optical devices.[1] Since the electronic and optical properties of quantum dots (QDs) are largely affected by the local potential environment around the dots, a precise understanding of the shape as well as the compositional profile of the dots is very important.[2–4] Investigation of the shape of the quantum dot itself can give many clues to the energetics and kinetic pathways involved in the dot formation. Moreover, studying the shape of uncapped SAQDs acts as a starting point for attaining precise control of QD geometry as well as understanding the morphological evolution of capped SAQD structures, which will be needed for device application.

In this regard, we first need to understand the shape of the uncapped SAQDs. There have been many reports of Ge(Si)/Si,[5–11] and III–V QD[12–22] morphology. In earlier reports on the InAs dots grown on GaAs by molecular beam epitaxy, detailed InAs dot shapes could not be resolved by atomic force microscopy (AFM) due to small island size and tip convolution effects.[12,15] Elongation of dots along the $[1\bar{1}0]$ direction was reported for $In_{0.3}Ga_{0.7}As$ quantum dots on GaAs,[16] and for InAs QDs on $In_{0.52}Al_{0.48}As/InP$.[23] {113},[16–18] and {136} faceting[13,19] has been proposed for InAs/GaAs quantum dots based upon *in situ* reflection high energy electron diffraction studies. Furthermore, {114} and {215} facets were also reported from a scanning tunneling microscopy study of InAs/GaAs dots.[18] High resolution transmission electron microscopy (HRTEM) has also been widely utilized to investigate the structures of QDs in detail.[19,24] In HRTEM studies, Georgsson *et al.* observed {110} and {111} facets in InP QDs on GaP/GaInP/GaAs,[24] and Lee *et al.* further showed the existence of {136} faceting in InAs/InP.[19] Recent reports on the morphology of InAs/InP QDs[14,23,25] reveal that the dots formed are slightly larger than in the InAs/GaAs system due to the reduced amount of misfit strain in this material system (~3.2%). AFM clearly showed the basal shape of these dots taking the form of a parallelogram,[14] whereas Lee *et al.* proposed a rhomboidal base.[13]

In this work, we performed a precise investigation on the shape of the InAs SAQDs grown on a lattice-matched InGaAs buffer layer on InP by AFM and TEM. We observed that the parallelogram dots were elongated either along $[1\bar{3}0]$ or $[\bar{3}10]$ directions. However, due to its unique geometry with {136} facets they looked as if they were overall elongated along $[1\bar{1}0]$ direction. We believe that under the growth conditions used in this investigation, the formation of these parallelogram dots is kinetically favored over rhombus dots, just as hut clusters can be kinetically favored over square pyramids in the SiGe/Si system.[5,6] The implication of this observation will be discussed, particularly regarding the evolution of III–V quantum dots.

The InAs SAQDs were grown on InGaAs/InP by MOCVD at 76 Torr. *Ex situ* cleansed and etched InP wafers were loaded into a horizontal quartz reactor. *In situ* oxide etching was performed in a PH_3+H_2 atmosphere at 620°C. 200-nm-thick InP and 2-nm-thick lattice-matched $In_{0.53}Ga_{0.47}As$ buffer layers were sequentially grown at 620°C and then the substrate temperature was ramped down to 550°C before the 2 monolayer (ML) growth of InAs. A V/III ratio of 30 was maintained during the QD growth, resulting in an InAs growth rate of 0.37 ML/s. Immediately following QD growth, the sample was cooled down to 300°C under an AsH_3+H_2 atmosphere, then to room temperature under a hydrogen atmosphere. The surface morphology of the InAs dots was measured by AFM (AutoProbe CP, PSIA) in contact mode, and bright field and high resolution TEM photographs were obtained using a JEOL JEM 2000 EX electron microscope at 200 kV.

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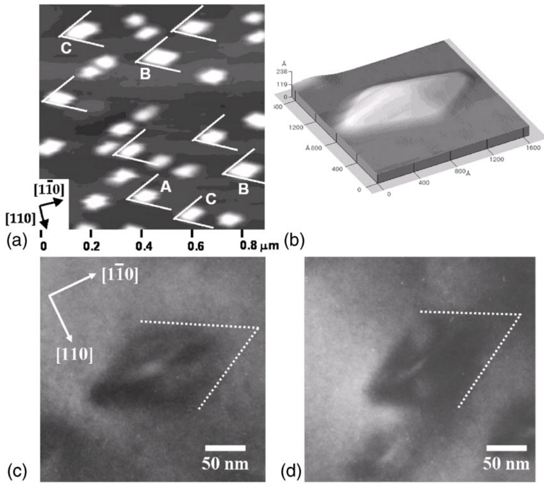

FIG. 1. (a) 0.9×0.9 μm AFM image of InAs SAQDs grown on $In_{0.53}Ga_{0.47}As/InP$ by MOCVD. The dot marked by "A" is a rhombus dot and the dots marked by "B" and "C" are two parallelogram dots elongated along $[1\bar{3}0]$ and $[\bar{3}10]$, respectively. (b) A magnified AFM image for a parallelogram QD "B." (c) and (d) are the TEM images for two parallelogram dots, "B" and "C," respectively.

Figure 1 shows a typical AFM image of InAs/InGaAs SAQDs (a), and a magnified image for an elongated dot (b). The areal dot density was 2.9×10^{9} cm^{-2}, and the average size of dots was about 131 nm (width along $[1\bar{1}0]$) $\times$13 nm (height). In Fig. 1(a), dots looked mostly elongated along $[1\bar{1}0]$, as often observed in III–V QD systems.[13,16–19,23] Close examination revealed that the dots took the shape of either a rhombus (marked "A") or a parallelogram (marked "B" and "C"). Island bases were aligned along $[1\bar{3}0]$ and $[\bar{3}10]$ directions and the chevrons in the image indicate these directions. The angle between the two lines of a chevron is 53.1° determined from the proposed dot shapes with {136} facets.[13,14] It is apparent that some dot facets were not fully developed. However, most dot facet baselines seem to be along $[1\bar{3}0]$ and $[\bar{3}10]$. Figures 1(c) and 1(d) are the plan-view TEM images for "B" and "C" type parallelogram dots, respectively. We found that the majority of the dots exhibited {136} faceting.

The facets and shape of dots were recognizable solely in the AFM image as shown in Fig. 1(b). But, for a geometrical point of view, AFM itself is not sufficient to describe a dot shape clearly due to tip convolution effects. To further investigate the {136} faceting behavior of these QDs, high-resolution cross-section TEM micrographs were taken along $[1\bar{1}0]$ (a) and [110] directions (b), as shown in Fig. 2. Angles between the (001) and vicinal planes were 25° and 13.3°, corresponding to {136} facet planes as shown in Fig. 2(c). These micrographs showed that the cross section of these dots takes a shape of a truncated triangle elongated along the $[1\bar{1}0]$ direction.

There are two elongation directions observed in these {136}-faceted dots. The first one is elongation along $[1\bar{1}0]$ direction forming {136} facets instead of isotropic {11n} facets. The second is along either $[\bar{3}10]$ or $[1\bar{3}0]$ directions resulting in parallelogram-base dots, rather than rhombus-base dots. The reason why the {136}-faceted dots were formed still remains in question. Surface dimers of III–V zinc-blende structure materials such as InP, GaAs, and InAs

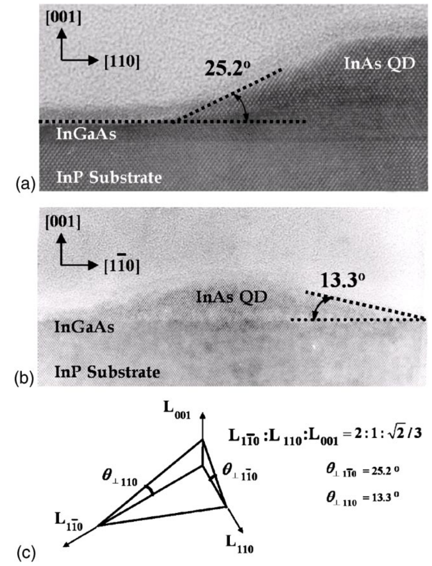

FIG. 2. High resolution XTEM micrographs either along $[1\bar{1}0]$ (a) and along [110] direction (b), respectively. Angles between (001) plane and the vicinal planes were 25° and 13.3°, as the theoretical calculations for {136} facet planes. (c) The geometry and the theoretical calculations for {136} faceted QD.

are well aligned along one direction for one atomic species. In GaAs, for example, (2×4) As dimers are aligned along $[1\bar{1}0]$ and (4×2) Ga dimers are along [110], resulting in anisotropic surface diffusion. So, considering that a non-strained III–V surface possesses an intrinsic anisotropy when As stabilized,[23] and that elongation along the $[1\bar{1}0]$ direction during the growth is usually observed in various III–V QD systems,[13,14,19,21,23] one can expect that elongation along $[1\bar{1}0]$ direction is energetically favorable.

Figure 3 shows three possible dot shapes incorporating {136} faceting. The major crystallographic orientations and the lateral dimensions of the dots, such as $L_{1\bar{1}0}$, L_{110}, $L_{\bar{3}10}$, and $L_{1\bar{3}0}$ are illustrated in Fig. 3. When the dots are symmetric along $[1\bar{1}0]$ and [110], they assume the shape of a rhombus[13] as shown in Fig. 3(a), where the ratio between $L_{1\bar{1}0}$ and L_{110}, $(L_{1\bar{1}0}/L_{110})$ is 2. A parallelogram-shaped island results when a rhomboidal island elongates along either $[\bar{3}10]$ or $[1\bar{3}0]$, while maintaining $L_{1\bar{1}0}/L_{110}$ at 2, as shown in

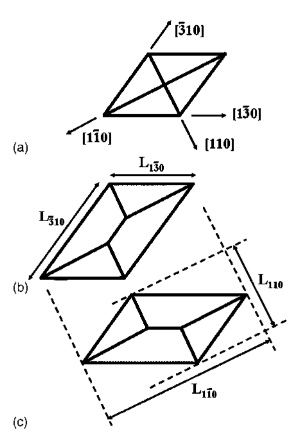

FIG. 3. Three types of InAs dots: (a) a rhombus dot, (b) a parallelogram dot elongated along $[\bar{3}10]$, and (c) a parallelogram dot elongated along $[1\bar{3}0]$. The important crystallographic orientations and the characteristic lengths are shown. The four planes constituting individual dots are {136} facets.

Figs. 3(b) and 3(c). Elongation in any other direction, such as along $[1\bar{1}0]$, leads to generation of facets other than {136}. Even though the dot shape proposed by Lee *et al.* was a rhombus, one of the XTEM photographs shown in Ref. [19] showed that the dot cross section was a truncated triangle, strongly suggesting that their dots were also elongated parallelograms.

Dot height (L_{100}) and width along [110] (L_{110}) are plotted against the lateral dimension along $[1\bar{1}0]$ ($L_{1\bar{1}0}$), as shown in Fig. 4. The straight line for L_{110} is a theoretical one expected from the parallelogram dot shape with {136} facets, with a slope of 0.5. The least-squares fitted slope from the data points is 0.504, in good agreement with the theoretical prediction. The slope of the L_{001} line (L_{100} vs $L_{1\bar{1}0}$) varies with the degree of island elongation, reaching a maximum for a rhombus-shaped dot. In this figure, a line with slope 0.12 (theoretical slope for a rhomboidal dot) is shown along with actual data. The least-squares fitted slope from the experimental data is 0.105, which is slightly smaller than 0.12, as expected, due to base elongation along $[\bar{3}10]$ or $[1\bar{3}0]$ directions.

In $Si_{1-x}Ge_x/Si$ QD systems, square pyramids and hut clusters were observed in the initial stage of dot evolution,[5–8] followed by the formation of higher aspect ratio domes with continued material deposition.[8,9] Square pyramids can elongate along elastically soft ⟨100⟩ directions, forming rectangular-base hut clusters, also bounded by {105} facets.[10] A kinetic barrier exists for the growth of strained facets,[5] which was found to be smaller for growth on smaller facets, leading to a shape transition from square base pyramids to rectangular base hut clusters as dots grow,[6] indicating that the growth of elongated islands (hut clusters) results from kinetic limitation. Even though the InAs dots are bounded with {136} facets and their shapes are different, a similar argument can be made for the observed rhombus to parallelogram shape changes. It is likely that the formation of parallelogram dots is also kinetically favored over larger isotropic rhombus dots having same volumes. Preferential growth induced by the smaller nucleation barrier of smaller facets may result in island elongation, especially in the case of kinetically limited dot growth.

Parallelogram dots, however, may transform to an energetically stable shape with or without further growth, in the absence of kinetic barriers at conditions close to equilibrium. The equilibrium shape of strained III–V quantum dots is not currently known. However, recent advances in SiGe/Si dot research[16,17,19] indicate that the shape would be of higher symmetry and with a higher aspect ratio (height/width). A report by Saito *et al.* on the shape transition of InAs dots from {136}-faceted to {110}-faceted dots as growth temperature varies from 510 to 550 °C strongly supports the fact that the equilibrium shape of III–V quantum dots is indeed composed of facets of higher symmetry.[21] The growth technique used in this case is alternating molecular beam epitaxy, which provides sufficient time for the near-equilibrium evolution of quantum dots.[16,22] In Fig. 1(a), some parallelogram dots have {110} facet segments normal to $[1\bar{1}0]$, suggesting that some of the dots are undergoing a shape transition toward higher symmetry shapes, consistent with the observations in Ref. [21].

We studied the shapes of InAs/InGaAs QDs by AFM and TEM. Parallelogram-base dots with {136} facets were

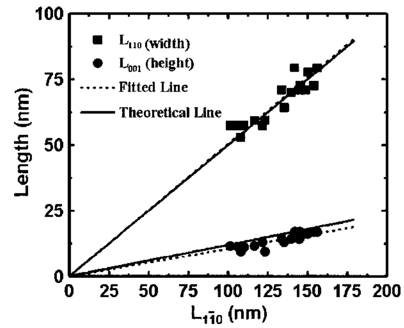

FIG. 4. Statistical analysis of {136}-faceted InAs dots. The data of L_{110} and L_{001} (height) are scattered and linear-plotted against $L_{1\bar{1}0}$. The solid line for L_{110} has a slope of 0.5, as predicted from the ideal {136}-faceted parallelogram dots. The solid line for L_{001} has a slope of 0.12 (the slope expected from rhombus dots) for visual aid.

observed with characteristic elongation directions along $[1\bar{3}0]$ and $[\bar{3}10]$. Some parallelogram dots with a small fraction of {110} facets were also observed and it is believed that they are undergoing a shape transition to dots of higher symmetry.

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